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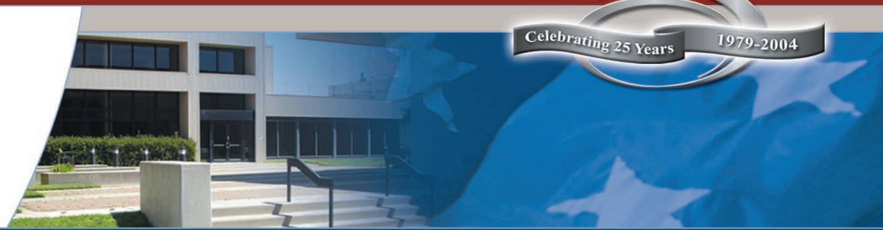
CRF NEWS



COMBUSTION RESEARCH FACILITY

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New Methods Produce Low-Temperature, Soot-Free Diesel Combustion

CRF researchers have discovered methods for producing non-sooting and low-flame temperature, diesel-like combustion. The research is motivated by the need to minimize nitrogen oxide (NO_x) and particulate matter (PM) emissions from high-efficiency diesel engines and is being conducted in collaboration with automotive and heavy-duty diesel manufacturers. The Department of Energy Office of FreedomCAR and Vehicle Technologies is sponsoring the research.

Lyle Pickett and Dennis Siebers conducted experiments at the CRF in reacting diesel fuel jets under well-controlled ambient and fuel injector conditions. The goal was to determine what factors affect soot formation during mixing-controlled diesel combustion with flame temperatures less than approximately 2000 K—too low for significant NO_x formation. The researchers showed that fuel jets that do not undergo soot formation in any region of the reacting jet, and that simultaneously have a low flame temperature, could be generated in at least three ways. The operating conditions and regions of heat release in the fuel jet (OH chemiluminescence images) for the three methods are shown in Figure 1. Simultaneously acquired planar laser-induced incandescence images (not shown) confirmed that the fuel jets were soot free.

A schematic illustrating how each method avoids soot formation and produces low flame temperatures is shown in Figure 2. The figure is an equivalence ratio versus temperature plot with contours indicating the locations where soot and NO_x formation occur for a diesel-like fuel. Curves (dashed lines) overlay the plot,

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Sandia National Laboratories

Quantifying Uncertainty

Researchers Develop Tools for Uncertainty Quantification in Computational Models of Reacting Flow

Confidence intervals on the predicted behavior of physical systems are necessary for optimization of engineering designs. They are also useful for model validation, which requires careful measurement of uncertainty in both experimental data and computational predictions. The goal of our uncertainty quantification (UQ) research is to evaluate the uncertainty in reacting flow model predictions resulting from uncertainties in parameters and other model inputs.

Habib Najm, Matthew Reagan, and Bert Debusschere of the CRF, along with Roger Ghanem and Omar Knio of the Johns Hopkins University and Olivier Le Maître of the Université d'Evry Val d'Essonne, have been developing tools for implementing UQ in a variety of applications. Studying chemical systems, they have found UQ to provide powerful analysis capabilities.

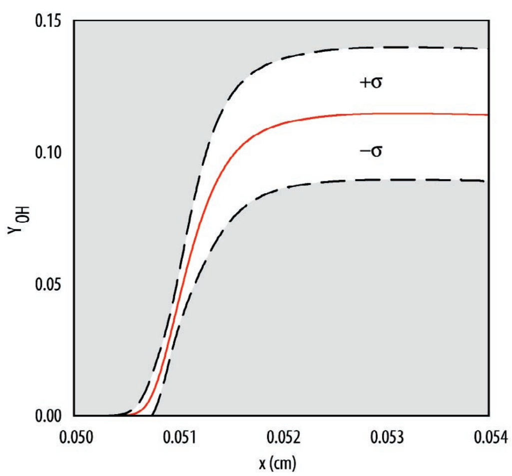

Figure 1. Profile of the OH mass fraction versus position in the planar premixed supercritical-water flame, with ±σ (standard deviation) bounds computed directly from the polynomial chaos expansion.

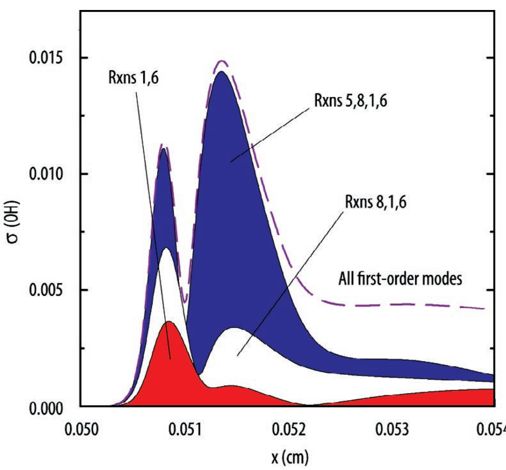

Figure 2. Standard deviation of OH mass fraction, showing the cumulative contribution of first-order dependencies on selected reactions and the total first-order contribution (reactions and enthalpies).

The simplest approach to UQ is through Monte Carlo (MC) simulations. For a more effective approach, a formalism has been developed using polynomial chaos (PC) expansions. In this formalism, each uncertain parameter is treated as a stochastic quantity with a known probability density function (PDF) and represented using a PC expansion. Using suitable means for propagating uncertainty through the model, its solution can be evaluated as a PC expansion. This can then be used to reconstruct the PDF of the solution.

These concepts can be implemented in different ways. The simplest case, "nonintrusive" spectral projection, involves generating deterministic realizations of the model predictions while sampling parameters from their known PDFs. A projection formalism allows one to compute the coefficients of the PC expansion of the solution from the set of individual realizations. While this approach is based on MC sampling, it provides a key advan-

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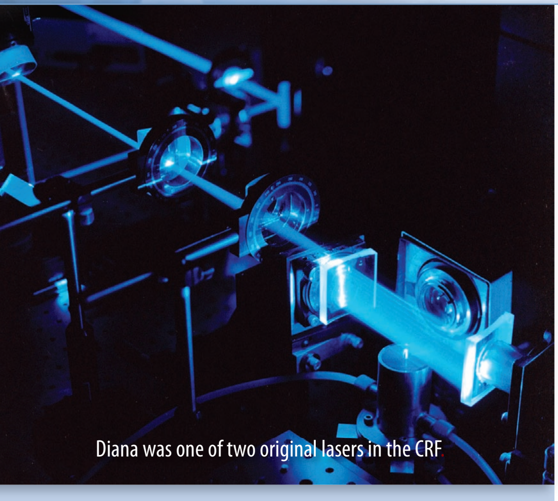


Diana was one of two original lasers in the CRF

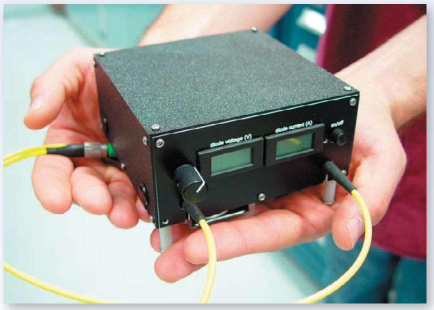
From Diana to the Fiber Amp

When the CRF opened to its first visitors in 1980, it housed two centralized lasers and a periscope distribution system. Diana was a lamp-pumped dye laser while Orion was a Nd:YAG slab glass laser developed by Sandia staff members in New Mexico. They were housed in a separate laser room at the end of the laboratory building. Overhead ducts with periscopes and relay mirrors were used to transmit the laser beams to labs.

However, a considerable amount of setup time was involved for each experiment. As commercial lasers improved, Diana and Orion were gradually phased out.

Today, the CRF houses about 200 high-powered lasers. CRF researchers largely use off-the-shelf equipment, which they frequently customize for specific purposes. Occasionally, they also forge new ground, as with the fiber laser amplifier.

Fiber lasers are attractive for a number of defense, manufacturing, and remote sensing applications because of their compact size, light weight, high electrical efficiency, and excellent beam quality. But low output power has been a limiting factor. Researchers at Sandia and the Naval Research Laboratory have invented a method that overcomes this limitation, allowing power to be scaled up by a factor of at least 100 while preserving beam quality and system efficiency. The advance promises to dramatically expand the potential applications of fiber lasers.



In contrast to Diana and Orion, a fiber laser and amp can almost fit in the palm of your hand.

Early Days of Using Lasers for Studying Combustion Were 'Exciting Time' for Sandia Researchers

Original 'Big' Lasers Now Replaced by Many Specialized, Ever-Shrinking Lasers

This article by CRF News editor Julie Hall continues our series commemorating the 25th anniversary of the CRF News and the CRF in 2005.

If the Combustion Research Facility were a company, it might appropriately be named "Lasers 'R Us."

While the CRF doesn't make or sell lasers, researchers use them in the majority of their work because of their ability to nonintrusively probe and gather data on fleeting chemical reactions and turbulent flows in hostile environments. This has been the CRF's charter and strength since the beginning. Over the past 25 years, it has developed an international reputation as a pioneer and leader in laser-based diagnostics, and is home to about 200 high-powered lasers, each performing a specialized role in the study of complex combustion phenomena.

Laser-based combustion research has changed significantly over that time. Gone are the days when the CRF housed two large lasers and piped the beams into individual labs. Mirroring the evolution of computers, today's lasers are increasingly smaller, cheaper, and more powerful. Computers and digital electronics have also proliferated in labs, allowing for measurements and data acquisition that previously would have been impossible using analog equipment.

Early laser work

The CRF's foundation in laser-based diagnostic methods was no accident. In the early 1970s, the nation plunged into the energy crisis, fueling widespread interest in improving energy efficiency.

At Sandia, researchers were using laser Raman scattering for weapons-related research on turbulent gas flow. A 1972 article published in *Science* reported the use of this technique for measurements in flames. The following year, Sandia began a program to develop laser-based diagnostics for combustion research.

Ron Hill and Dan Hartley observe their light trapping cell in operation.

"Initial flame experiments at Sandia, facilitated by unique multi-pass optics to enhance laser power, helped launch the Sandia effort towards the forefront of applying laser diagnostics to combustion studies," said Bob Setchell, an early Sandia combustion researcher, now working in Sandia's Radiation-Solid Interactions Department.

When the CRF welcomed the first users in 1980, "laser diagnostics became the heart of the center," said Pete Witze.

Lasers in engines

Witze was part of the research team that in 1979 first applied laser diagnostics to an internal combustion engine. In those days, automotive engineers did not have expensive tools like lasers. The Sandia researchers made the first reported measurements in an optically modified, single-cylinder engine using the most powerful continuous-wave argon-ion laser that was commercially available at the time.

It was a significant advance that would lead to a strong collaboration with the automotive industry that continues today.
Because using lasers to study combustion was so new, breakthroughs were rather commonplace.
"No matter what you did, nobody had done anything in that area. There were all these 'firsts'," Witze said.

"It was a very exciting and very innovative

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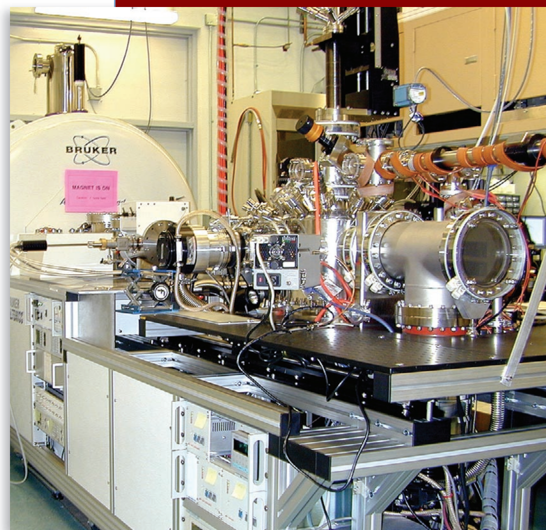
Sandia Receives National Institutes of Health Grant to Construct Data Portal for Biomedical Researchers

The National Institutes of Health has awarded a five-year, $2.5 million grant to Sandia to construct a data portal for the biomedical research community. The project, which is funded jointly by the National Science Foundation, involves researchers in the CRF and other organizations within Sandia, as well as the University of California, San Francisco, and the University of Maryland, Baltimore County.

The team will construct a knowledge grid for the biomedical community with secure data/metadata management, application integration, collaboration tools, analysis capabilities, and research tools. This work will be carried out in direct collaboration with scientists leading the development of MS3D, a new method that combines intramolecular chemical crosslinking with high-resolution mass spectrometry to glean structure information about proteins and other biological macromolecules. The Collaboratory for MS3D will pilot the integration and evaluation of these tools and monitor the impact of the resulting knowledge grid on a newly developing scientific community.

Work on the new collaboratory is a direct outgrowth of the CRF's Collaboratory for Multi-scale Chemical Science (CMCS) **(http://cmcs.org)** and will build on technology developed through that project. CMCS is a Department of Energy-funded project designed to break down barriers to rapid sharing of validated chemical science information and open new paradigms for collaborative science. "Collaboratories" link geographically dispersed researchers, data, and tools via high-performance networks to enable remote access to facilities, access to large datasets, shared environments, and ease of collaboration.



The chemical microprobe Fourier-transform ion cyclotron resonance mass spectrometer, located in the CRF, will be used to provide data for the MS3D project.

Massachusetts Institute of Technology Doctoral Graduate Comes to CRF Through New Sandia Fellowship Program

Youssef Marzouk, a Massachusetts Institute of Technology (MIT) graduate, joined the Reacting Flow Research Department in September as one of the first recipients of the President Harry S. Truman Research Fellowship in National Security Science and Engineering. Marzouk will be mentored by Larry Rahn, and will work with Habib Najm on Bayesian inference for inverse problems and optimization, with applications in fluid dynamics, source inversion, and gene regulatory networks.

Marzouk and Gregory Nielson, also of MIT, were selected for the new Sandia postdoctoral fellowship program after an intensive 10-month national search. Nielson will work at Sandia's New Mexico site.

The Truman Fellowship gives recipients the opportunity to pursue independent research of their own choosing for three years that supports Sandia's national security mission. Fellowship candidates are expected to have solved a major scientific or engineering problem in their thesis work or to have provided a new approach or insight to a major problem, as evidenced by a recognized impact in their field.

Marzouk received his bachelor's, master's, and Ph.D. degrees from MIT. He was the recipient of the Fannie and John Hertz Foundation Graduate Fellowship. He received the Young Researcher Fellowship Award at the MIT Conference on Computational Fluid and Solid Mechanics and was the recipient of the Barry Goldwater Scholarship. Recently, he was awarded the Joseph H. Keenan Prize for outstanding graduate student in the thermal sciences at MIT. His research experience includes work at the MIT Reacting Gas Dynamics Laboratory and the MIT Fluid Mechanics Laboratory.

Gregory Fiechtner, a staff member in Sandia's Microfluidics Department, has taken a two-year position with the Molecular Processes and Geosciences Team in the Office of Science, Basic Energy Sciences, at the Department of Energy. His primary responsibility will be the separations and analysis portfolio.

Jerry Caton, a mechanical engineering professor at Texas A&M University, left the CRF in August. Since May, Caton had been working with Sandians Lyle Pickett and Mark Musculus on modeling diesel combustion with detailed chemistry.

Former CRF postdoc Petr Novak (right) has joined the technical staff of the Institute of Microbiology at the Academy of Sciences of the Czech Republic. He had worked at the CRF for two years on the Interfacial Bioscience Grand Challenge (IBIG), a project that harnesses unique experimental and computational capabilities to achieve a better understanding of communication across cell membranes via protein interactions.

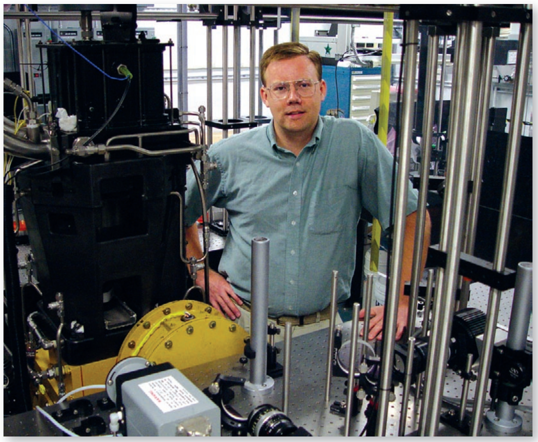
Postdoc Will Haskins (not pictured) also left the CRF recently to take an associate scientist position at Genentech. He had worked on the IBIG project for about eight months. Haskins had been involved in research on the three-dimensional structure of proteins using chemical crosslinking and Fourier-transform mass spectrometry.

Three Win Society of Automotive Engineers Presentation Awards

Chuck Mueller, Pete Witze, and Paul Miles of the CRF recently received Society of Automotive Engineers (SAE) Awards for Excellence in Oral Presentation for papers presented at two different conferences.

Mueller received two awards for presentations given at the June 2004 SAE Fuels and Lubricants Meeting and Exhibition in Toulouse, France. One presentation, summarizing results from a collaboration between Sandia and Caterpillar Inc., described how dual-injection operating conditions that employ early direct-injection homogeneous charge compression ignition combustion of diesel fuel affect in-cylinder processes and engine-out emissions. The other presentation, on results from a collaboration between Sandia and Lawrence Livermore National Laboratory, described the use of carbon-14 isotope labeling and tracing to show how the molecular structure of a diesel oxygenate can influence its ability to lower engine-out soot emissions.

Witze and Miles received their awards for presentations at the March 2004 SAE International Congress and Exposition in Detroit. Witze's presentation on laser-induced incandescence (LII) measurements of diesel particulate emissions detailed results from a collaboration performed at the Ford Vehicle Emissions Research Laboratory comparing LII measurements with several other techniques for the light-duty Federal Test Procedure. In Miles' presentation, he identified the dominant sources of in-cylinder turbulence production in a high-speed diesel engine, and clarified the influence of flow swirl on the various turbulence sources. This combined experimental and numerical study was a collaboration between Sandia, the University of Wisconsin, Wayne State University, and the University of Michigan.



University of Illinois student Glen Martin completed his Ph.D. in mechanical engineering in September after successfully defending his thesis. Working with Sandian Chuck Mueller, Martin has been conducting his Ph.D. research for the past three years in the Liquid Fuels Research Lab.

New Methods *(Continued from page 1)*

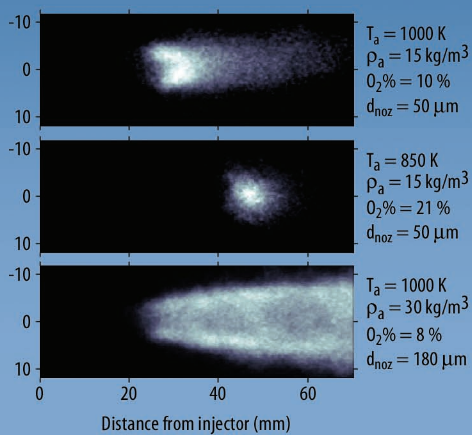


Figure 1. OH chemiluminescence images of three methods for low-flame temperature, soot-free diesel combustion. Images are time-averaged during mixing-controlled heat release phase of combustion. The fuel injector is at the image's left edge. Ambient temperature, density, oxygen concentration, and injector nozzle size are listed to the right. The injector orifice pressure drop was 1400 bar.

showing the predicted adiabatic flame temperatures for fuel-air mixtures at the given equivalence ratios for four ambient oxygen concentrations. The reduced oxygen concentrations simulate the effect of various levels of exhaust-gas recirculation (EGR) in an engine.

The first method (1) uses a small nozzle coupled with a low ambient oxygen concentration. The small nozzle induces fast fuel-air mixing prior to the flame lift-off length to produce equivalence ratios less than two in the fuel jet prior to combustion, thereby avoiding soot-forming rich fuel-air mixtures. The ambient gas oxygen concentration of 10% keeps stoichiometric flame temperatures below 2000 K, as indicated by the solid portion of the adiabatic flame temperature curve. The second method (2) uses a small nozzle coupled with a cooler ambient temperature to extend the lift-off length and induce more fuel-air mixing prior to the lift-off length, resulting in an equivalence ratio of approximately 0.6 prior to combustion. The lean mixture avoids both soot formation and a high-temperature stoichiometric diffusion flame. The third method (3) relies on the use of very high EGR (8% ambient oxygen), resulting in mixtures in the fuel jet that are rich, but with peak adiabatic flame temperatures that are too cool for soot inception at diesel timescales.

The lack of soot formation and low flame temperatures realized in the reacting fuel jets suggest that these diesel combustion methods offer the potential for a simultaneous soot and NO_x reduction in an engine, while maintaining a mixing-controlled heat release rate. Although much further research is needed to determine if these non-sooting, low-temperature mixing-controlled combustion strategies can be employed in engines, the results are providing guidance to engine designers on directions to proceed to lower soot and NO_x emissions.

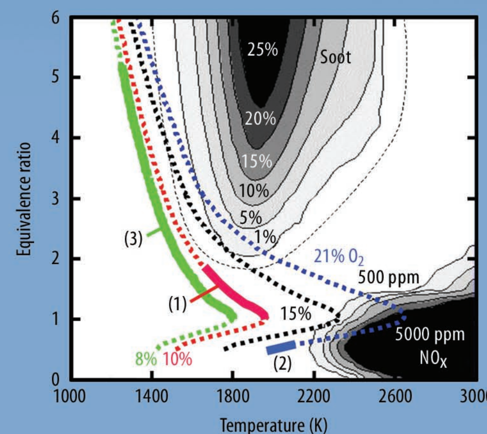


Figure 2. Schematic of equivalence ratio versus adiabatic mixture flame temperature.

Using Lasers for Studying Combustion *(Continued from page 2)*

time," said Marshall Lapp, who coauthored the 1972 *Science* article while at General Electric and later became a Sandia employee.

Laser diagnostic milestones

For about two decades, the Department of Energy Office of Basic Energy Sciences has funded a highly successful diagnostics research program at the CRF. Through that program, CRF researchers have made many significant contributions to the science of laser diagnostics, including the following developments or milestones:

• Some of the first optically based methods to detect atomic hydrogen in a combustion environment;

• Detailed energy transfer and quenching studies and associated mod els that allow quantitative interpretation of laser-induced fluorescence;

• Detailed lineshape studies, intensity effects, and models that allow quantitative interpretation of coherent anti-Stokes Raman spectroscopy (CARS);

• Degenerate four-wave mixing techniques and models;

• Nondegenerate four-wave mixing techniques and model descriptions;

• Injection-seeding approaches for lasers that are in widespread use in commercial lasers for combustion measurements;

• Significant enhancements of the spatial resolution and sensitivity of CARS for use in combustion studies;

• Measurement of ultrashort laser pulses—a difficult problem—using a technique called frequency-resolved optical gating;

• Development of a picosecond laser that offers a unique combination of time and spectral resolution and enables a detailed understanding of collisional phenomena;

• Developed ion imaging, a way of measuring the velocity of slowly moving ions and gaining information about bond strengths and velocities of the reactions products.

Uncertainty Quantification (Continued from page 1)

tage over MC by enabling the identification of the contribution of individual parameters to the uncertainty in model predictions. This method is simple to implement but requires a large number of deterministic realizations to converge.

PC UQ analysis can also be done "intrusively" by reformulating the governing equations into corresponding equations for the PC expansion coefficients of the solution and solving for these directly. This method demands the development of specialized numerical algorithms and computational code to solve the reformulated equation system, but total computing time can be significantly reduced, depending on the properties of the governing equations.

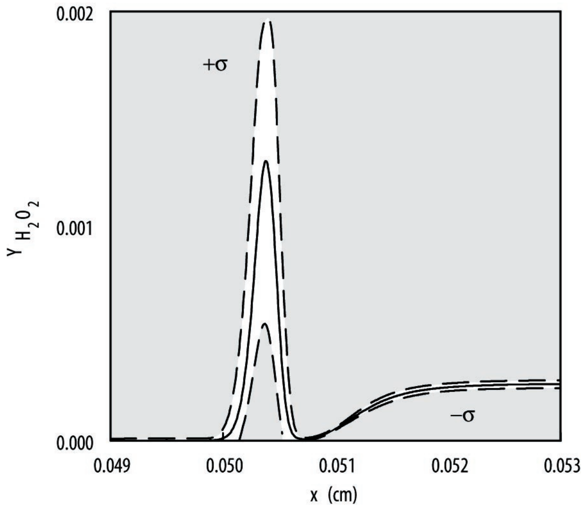


Figure 3. Profile of H_2O_2 mass fraction versus position in the planar premixed supercritical-water flame, with ±σ bounds.

The researchers used the sampling-based, nonintrusive, PC construction for a UQ study of a planar premixed flame front burning into a supercritical hydrogen-oxygen mixture. The model accounted for known uncertainties in the eight reaction rate pre-exponential constants and five standard-state heats of formation. Parametric samples were generated using a Latin hypercube sampling strategy, and model realizations were solved using the Sandia PREMIX code. Resulting statistics were processed and projected onto the PC modes of the model output fields.

Figure 1 shows the spatial distribution of the mean and standard deviation of the OH radical mass fraction. The figure presents a close-up of the flame region, with reactants on the left and products on the right. The mean OH field exhibits the expected fast rise of OH in the reaction zone. The standard deviation envelope illustrates the fast amplification in uncertainty in the reaction zone, leading to large OH uncertainty in the post-flame region.

Figure 2 illustrates the technique's ability to highlight contributions of individual parameters to uncertainty in the OH field. The figure shows the cumulative first-order contributions of each uncertain parameter to the standard deviation of OH. The results highlight the relative contributions of different reactions to the resulting OH uncertainty at different points in the flame, indicating clearly the large contribution due to Rxn. 5 in the model (H_2O_2+OH = H_2O + HO_2). This information is useful for guiding subsequent experimental studies. Clearly, reducing the uncertainty in the rate constant of this reaction, through improved measurements, will have maximal impact on reducing uncertainty in predicted OH with this model.

Another key observation in the results is the large uncertainty evident in some of the radical fields. Specifically, H_2O_2 predictions exhibit a large standard deviation in the flame zone, as seen in Figure 3. The uncertainty in H_2O_2 is large enough to raise questions about the robustness of the present model for predictions of this flame reaction zone radical under the given conditions. Moreover, it suggests that H_2O_2 would not be the optimal choice of flame observable for validating this model under these conditions.

Sandia National Laboratories

Mail Stop 9052
P.O. Box 969
Livermore, California 94551-0969

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Editor .... Julie Hall
Graphic Artist .... Daniel Strong
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Subscriptions are free. Subscriptions and address changes to Julie Hall, jchall@sandia.gov.

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